SUPPLEMENTARY MATERIAL

Fossil versus contemporary sources of fine elemental and organic carbonaceous particulate matter during the DAURE campaign in Northeast Spain

M.C. Minguillón[1,2,*], N. Perron[1, #], X. Querol[2], S. Szidat[3,4], S.M. Fahrni[1,3,4], A. Alastuey[2], J.L. Jimenez[5,6], C. Mohr[1], A.M. Ortega[5,7], D.A. Day[5,6], V.A. Lanz[1], L. Wacker[8], C. Reche[2], M. Cusack[2], F. Amato[2], G. Kiss[9], A. Hoffer[9], S. Decesari[10], F. Moretti[10], R. Hillamo[11], K. Teinilä[11], R. Seco[12,+], J. Peñuelas[12], A. Metzger[13], S. Schallhart[13], M. Müller[13], A. Hansel[13], J.F. Burkhart[14], U. Baltensperger[1], A.S.H. Prévôt[1]

[1]Paul Scherrer Institut, Laboratory of Atmospheric Chemistry, 5232 Villigen PSI, Switzerland
[2]Institute of Environmental Assessment and Water Research (IDAEA), CSIC, 08034 Barcelona, Spain
[3]Dept. of Chemistry and Biochemistry, University of Bern, 3012 Berne, Switzerland
[4]Oeschger Centre for Climate Change Research, University of Bern, 3012 Berne, Switzerland
[5]Dept. of Chemistry and Biochemistry, University of Colorado, Boulder, CO, USA
[6]Cooperative Institute for Research in the Environmental Sciences (CIRES), University of Colorado, Boulder, CO, USA
[7]Dept. of Atmospheric and Oceanic Science, University of Colorado, Boulder, CO, USA
[8]Laboratory of Ion Beam Physics, ETH Hönggerberg, 8093 Zurich, Switzerland
[9]Air Chemistry Group of the Hungarian Academy of Sciences, H-8201 Veszprém, Hungary
[10]Institute for Atmospheric Sciences and Climate (ISAC), CNR, Bologna, Italy
[11]Finnish Meteorological Institute, Air Quality Research, 00101 Helsinki, Finland
[12]Unitat d'Ecologia Global CREAF-CEAB-CSIC, Centre de Recerca Ecològica i Aplicacions Forestals (CREAF), Universitat Autònoma de Barcelona, 08193 Bellaterra, Barcelona, Spain
[13] Institute of Ion Physics and Applied Physics, Innsbruck University, A-6020 Innsbruck, Austria
[14]Norwegian Institute for Air Research (NILU), NO-2027 Kjeller, Norway
[#] now at Dept. of Nuclear Physics, University of Lund, 221 00 Lund, Sweden
[+] now at Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, CO 80301, USA

*Correspondence to: M.C. Minguillón (mariacruz.minguillon@idaea.csic.es)

SECTION S1

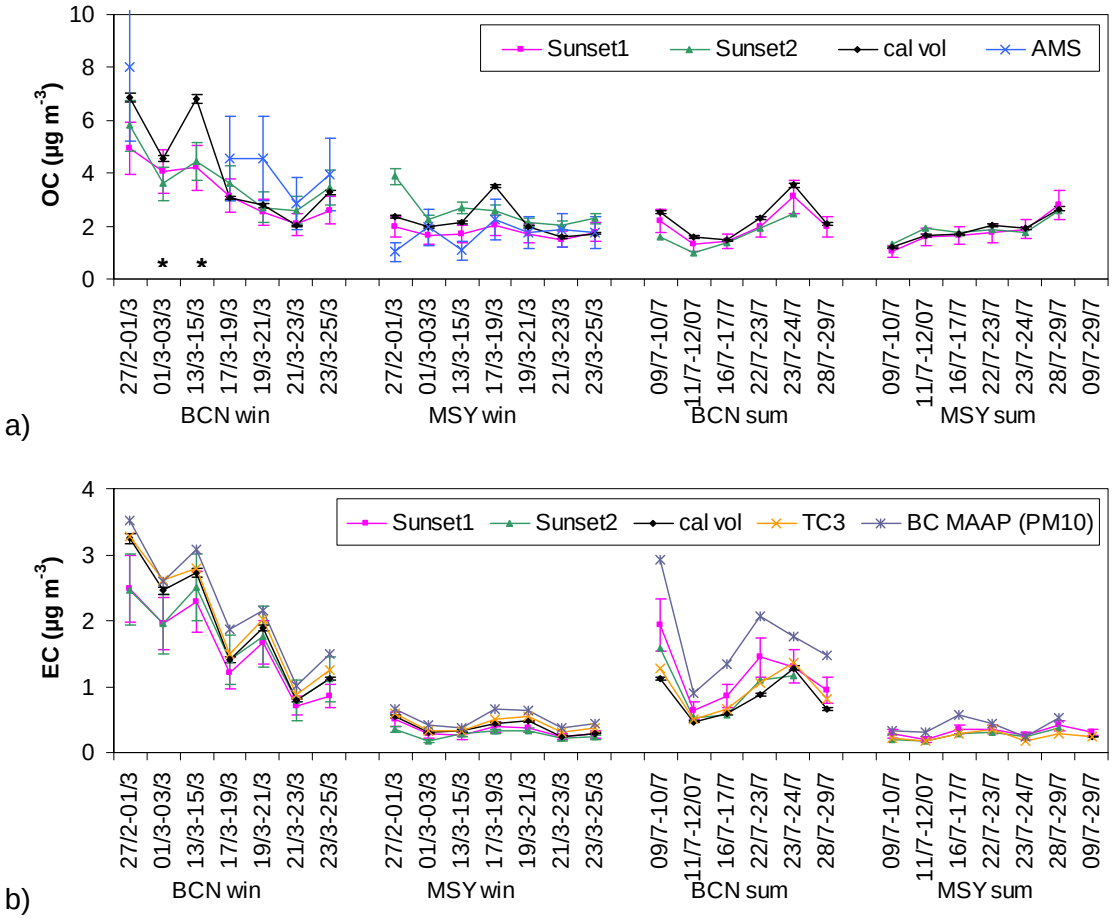

Figure S1. a) Organic carbon (OC) concentrations in PM_1 measured by Sunset1, Sunset2 and calibrated volume (cal vol); and calculated from organic matter measured by AMS using the AMS-determined OM/OC ratios of the different OA components for BCN and using the OM/OC ratios determined by AMS for MSY . *: Low availability of AMS data, so average not plotted.

b) Elemental carbon (EC) concentrations in PM_1 measured by Sunset1, Sunset2 and calibrated volume (cal vol); and measured by the Sunset instrument during the third stage of thermal program for EC collection (TC3); and BC concentrations in PM_{10} measured by MAAP.

SECTION S2

Table S1. Compilation of literature values of the EC/OC and levoglucosan/OC ratios for biomass burning emissions. SW: softwood; HW hardwood.

Reference	Notes	$(EC/OC)_{bbe}$	unc $(EC/OC)_{bbe}$	$(lev/OC)_{bbe}$	unc $(lev/OC)_{bbe}$
Bond et al., 2004		0.161			
Chow et al., 2010	residencial wood comb	0.188			
Chow et al., 2010	agricultural burning	0.300			
Chow et al., 2010	open/prescribed burning	0.180			
Chow et al., 2010	forest fire	0.043			
Chow et al., 2010	wildfires	0.513			
Chow et al., 2010	fires	0.513			
Fine et al. (2002, 2004)	Pine			0.258	0.052
Fine et al. (2002, 2004)	Region 5 profile			0.146	0.029
Fine et al. (2002, 2004)	Region 4 profile			0.093	0.019
Fine et al., 2004	Quaking Aspen	0.016	0.003	0.188	
Fine et al., 2004	Douglas Fir	0.054	0.007	0.271	
Fine et al., 2004	Ponderosa Pine	0.081	0.009	0.071	
Fine et al., 2004	Pinyon Pine	0.408	0.032	0.01	
Fine et al., 2004	White Oak	0.015	0.003	0.098	
Fine et al., 2004	Sugar Maple	0.044	0.006	0.168	
Fine et al., 2004	Black Oak	0.030	0.004	0.234	
Fine et al., 2004	American Beech	0.015	0.003	0.076	
Fine et al., 2004	Black Cherry	0.022	0.006	0.334	
Fine et al., 2004	White Spruce	0.039	0.005	0.142	
Gonçalves et al., 2010	Eucalyptus globulus HW	0.32		0.462	
Gonçalves et al., 2010	Pinus pinaster SW	1.11		0.146	
Gonçalves et al., 2010	Quercus suber HW	0.23		0.159	
Gonçalves et al., 2010	Acacia longifolia HW	1		0.096	
Liousse et al., 1996		0.139			
McMeeking et al., 2009	Montane	0.022			
McMeeking et al., 2009	Douglas fir	0.014			
McMeeking et al., 2009	Lodgepole pine	0.040			
McMeeking et al., 2009	Ponderosa pine	0.027			
McMeeking et al., 2009	Rangeland	0.128			
McMeeking et al., 2009	Juniper	3.857			
McMeeking et al., 2009	Rabbitbrush	2.800			
McMeeking et al., 2009	Sagebrush	0.041			
McMeeking et al., 2009	Chaparral	0.076			
McMeeking et al., 2009	Ceanothus	0.092			
McMeeking et al., 2009	Chamise	0.175			
McMeeking et al., 2009	Manzanita	0.024			
McMeeking et al., 2009	Coastal plain	0.073			
McMeeking et al., 2009	Black needlerush	0.016			
McMeeking et al., 2009	Common reed	0.020			
McMeeking et al., 2009	Gallberry	1.141			
McMeeking et al., 2009	Hickory	0.042			
McMeeking et al., 2009	Kudzu	0.014			
McMeeking et al., 2009	Longleaf pine	0.039			
McMeeking et al., 2009	Oak	0.038			
McMeeking et al., 2009	Palmetto	0.094			
McMeeking et al., 2009	Rhododendron	0.095			
McMeeking et al., 2009	Sawgrass	0.120			

Reference	Notes	EC/OC	unc EC/OC	lev/OC	unc lev/OC
McMeeking et al., 2009	Turkey oak	0.043			
McMeeking et al., 2009	Wax myrtle	0.056			
McMeeking et al., 2009	Wire grass	0.086			
McMeeking et al., 2009	Boreal forest	0.026			
McMeeking et al., 2009	Alaskan duff	0.000			
McMeeking et al., 2009	Black spruce	0.097			
McMeeking et al., 2009	White spruce	0.037			
McMeeking et al., 2009	Other	160			
McMeeking et al., 2009	Fern	0.045			
McMeeking et al., 2009	Rice straw	194			
Saarikoski et al., 2008		0.152			
Saarnio et al., 2010	plume measurements	0.100	0.068		
Saarnio et al., 2010	plume measurements	0.106	0.044		
Saarnio et al., 2010	plume measurements	0.149	0.068	0.028	0.014
Sandradewi et al., 2008		0.137			
Schmidl et al., 2008	Beech	0.372		0.080	
Schmidl et al., 2008	Oak	0.308		0.273	
Schmidl et al., 2008	Spruce	0.384		0.199	
Schmidl et al., 2008	Larch	0.176		0.272	
Schmidl et al., 2008	Briquettes	0.760		0.248	
Sullivan et al., 2008	Alaskan Duff			0.117	
Sullivan et al., 2008	Black Needle Rush	0.039		0.080	
Sullivan et al., 2008	Black Spruce	0.312		0.073	
Sullivan et al., 2008	Black Spruce, dried	0.087		0.083	
Sullivan et al., 2008	Black Spruce, fresh	0.010		0.079	
Sullivan et al., 2008	Ceanothus	0.098		0.053	
Sullivan et al., 2008	Chamise	0.668		0.065	
Sullivan et al., 2008	Fir			0.049	
Sullivan et al., 2008	Gallberry	1.068		0.033	
Sullivan et al., 2008	Grass	0.050		0.030	
Sullivan et al., 2008	Hickory	0.043		0.037	
Sullivan et al., 2008	Juniper	3.455		0.015	
Sullivan et al., 2008	Kudzo			0.025	
Sullivan et al., 2008	Lodgepole Pine Needle Duff			0.134	
Sullivan et al., 2008	Lodgepole Pine, dead/small	1.042		0.186	
Sullivan et al., 2008	Lodgepole Pine, fresh	0.005		0.053	
Sullivan et al., 2008	Longleaf Pine	0.042		0.072	
Sullivan et al., 2008	Manzanita	0.293		0.072	
Sullivan et al., 2008	Oak	0.038		0.062	
Sullivan et al., 2008	Palmetto	0.364		0.055	
Sullivan et al., 2008	Phragmites	0.019		0.075	
Sullivan et al., 2008	Ponderosa Pine Duff	0.009		0.069	
Sullivan et al., 2008	Ponderosa Pine, dead	0.790		0.102	
Sullivan et al., 2008	Ponderosa Pine, fresh			0.070	
Sullivan et al., 2008	Puerto Rican Fern	0.058		0.070	
Sullivan et al., 2008	Puerto Rican Mixed Woods	0.104		0.129	
Sullivan et al., 2008	Rhododendron	0.101		0.101	
Sullivan et al., 2008	Rice Straw (Taiwan)	0.039		0.076	
Sullivan et al., 2008	Sage	0.023		0.028	
Sullivan et al., 2008	Saw Grass	0.124		0.041	
Sullivan et al., 2008	Southern Pine, dried	0.068		0.098	
Sullivan et al., 2008	Titi	0.343		0.051	
Sullivan et al., 2008	Turkey Oak	0.045		0.047	

Reference	Notes	EC/OC	unc EC/OC	lev/OC	unc lev/OC
Sullivan et al., 2008	Wax Myrtle	0.131		0.056	
Sullivan et al., 2008	Wax Myrtle	0.038		0.059	
Sullivan et al., 2008	White Spruce	0.000		0.133	
Sullivan et al., 2008	Wiregrass	0.066		0.201	
Sullivan et al., 2008	Wiregrass	0.098		0.172	
Szidat et al., 2009		0.157	0.05	0.1	0.08

Table 3 Szidat et al., 2006, JGR					
Edgerton et al.1986		0.175			
Rau, 1989		0.163			
Hildemann et al.1991		0.084			
Cachier et al.1996		0.116			
Liousse et al.1996		0.234			
McDonald et al.2000		0.182			
Fine et al.2001		0.153		0.103	
Schauer et al.2001		0.04		0.246	
Fine et al.2002		0.159		0.042	
Fine et al. 2004a		0.086		0.134	
Fine et al. 2004b		0.213		0.245	
AVE Szidat et al., 2006		0.16	0.05	0.15	0.09

Table 2 Reid et al., 2005, ACP					
Andreae et al. 1998	Savanna	0.23			
Cachier et al. 1995	Savanna	0.12			
Cachier et al. 1996	Savanna	0.12			
Ferek et al. 1998	Cerrado	0.12			
Ferek et al. 1998	Grass/Pasture	0.11			
Formenti et al. 2003	Savanna	0.06	0.01		
Liousse et al. 1995	Savanna	0.13	0.01		
Ward et al. 1992	Cerrado	0.06			
Hobbs et al. 1996	Presc. Temperate	0.08	0.02		
Mazurek et al. 1991	Boreal	0.07	0.02		
Susott et al. 1991	Temperate	0.15			
Ward et al. 1992	Temperate	0.1			
Mazurek et al. 1991	Boreal Forest	0.08	0.03		
Mazurek et al. 1991	Boreal Forest	0.03	0.03		
Susott et al. 1991	Temperate	0.04			
Ferek et al. 1998	S. Amer	0.15			
Ferek et al. 1998	S. Amer	0.08			
Ward et al. 1992	S. Amer	0.2			
Patterson, 1984	Forest Debris	0.6			
Turn et al. 1997	Herbatious Fuel	0.52			
Turn et al. 1997	Woody Fuel	0.48			

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Schmidl, C., Marr, I.L., Caseiro, A., Kotianová, P., Berner, A., Bauer, H., Kasper-Giebl, A., and Puxbaum, H.: Chemical characterisation of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, Atmos. Environ., 42, 126-141, 2008.

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Szidat, S., Jenk, T. M., Synal, H.-A., Kalberer, M., Wacker, L., Hajdas, I., Kasper-Giebl, A., and Baltensperger, U.: Contributions of fossil fuel, biomass-burning, and biogenic emissions to carbonaceous aerosols in Zurich as traced by ^{14}C. J. Geophys. Res.-Atmos., 111, D07206, doi:10.1029/2005JD006590, 2006.

Szidat, S., Ruff, M., Perron, N., Wacker, L., Synal, H. A., Hallquist, M., Shannigrahi, A. S., Yttri, K. E., Dye, C., and Simpson, D.: Fossil and non-fossil sources of organic carbon (OC) and elemental carbon (EC) in Göteborg, Sweden, Atmos. Chem. Phys., 9, 1521-1535, 2009.

SECTION S3

New method for EC collection

The new method for EC collection for correct ^{14}C determination (still under development, Zhang et al., 2011) consists of coupling a Sunset Instrument to the cryo-trap system (as opposed to an oven with a fixed temperature with the old method), so that the thermal cycles can be defined accurately. The whole thermal program is carried out under pure oxygen. The CO_2 evolving from the 'EC step' is trapped for subsequent ^{14}C analysis. The filter transmittance is monitored continuously by the attenuation of a laser, which is used to calculate the EC yields in the CO_2 collection stage. The thermal program used consists on a first step at 400ºC (T1) during 150 seconds (including pre-step 1), a cooling time of 18 seconds, a second step at 500ºC (T2) during 75 seconds, another cooling during 40 seconds, and a third step at 760ºC (T3) during 150 seconds, named 'EC step', during which the CO_2 evolved is collected. All these temperatures are the set temperatures with the real thermal evolution being different (Figure S2). The second step in this method should be optimized with the goal of a complete removal of the carbonaceous interfering fraction with the best possible recovery of the elemental carbon. To this end, different temperatures for this second step (T2) need to be tested and results evaluated (Figure S2).

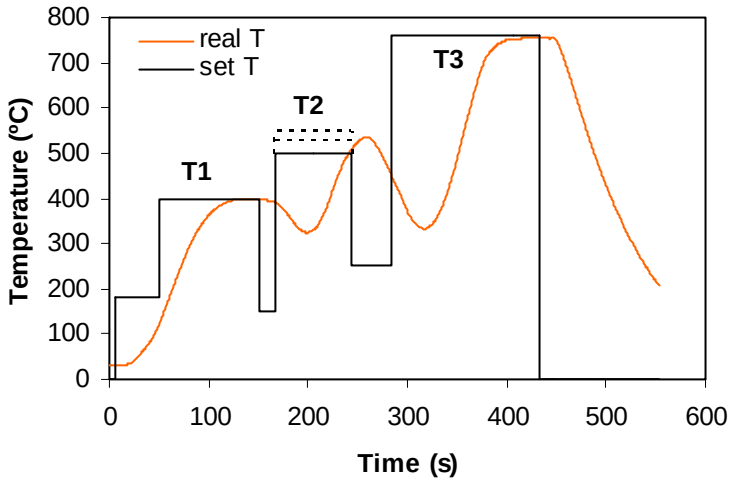

Figure S2. Thermal program on the Sunset instrument for the EC collection for subsequent ^{14}C analysis during the third step (at T3), showing the variation of T2 for optimizing tests.

References from Section S3:

Zhang, Y.L., Perron, N., Zotter, P., Minguillón, M.C., Prévôt, A.S.H., Wacker, L., and Szidat, S.: On the isolation of organic carbon and elemental carbon of carbonaceous aerosols for 14C measurement: A modified thermal/optical method, in preparation, 2011.

SECTION S4

Selection of T2 for the EC collection thermal program

Different temperatures ranging from 485 to 600ºC (as set T2 temperatures) were tested using two filters from the study campaign, one from Barcelona (S5452) and one from Montseny (S5943). The yields of EC in the third step are defined by the attenuation (ATN) of the laser signal, hence the ratio ATN/ATNi (i standing for initial) indicates the amount of EC with respect to the initial (total) amount of EC in the filter. Figure S3 shows the ATN/ATNi right before step 3 for different T2. The EC yield does not change significantly when increasing the T2 up to around 560ºC (real temperature). That indicates that the amount of carbon removed at the second step does not change, hence, any temperature chosen for T2 in this range would be valid, without any further information.

Additionally, analyses of fM of the collected CO_2 during the third step were carried out for different tests using different T2 (Figure S4).

The filter from Barcelona shows a slight decrease in the fM when increasing the T2, nevertheless the EC yield does not change, as explained above, this leading to the conclusion that the variation in the fM is not significant.

The filter from Montseny does not show any trend in the fM when increasing the T2 from 530 to 560 ºC, and the EC yield didn't change significantly, as explained above.

Hence, for the type of samples of this study, the thermal program used allows us to get EC yields over 80% and the fM obtained from these yields is consistent and therefore it is considered representative of the EC collected and one can assume that it is representative of the whole EC fraction. The selection of the T2 could then be any T within the tested range, and the T2 was then set at 500ºC (set temperature, corresponding to 530ºC as real temperature).

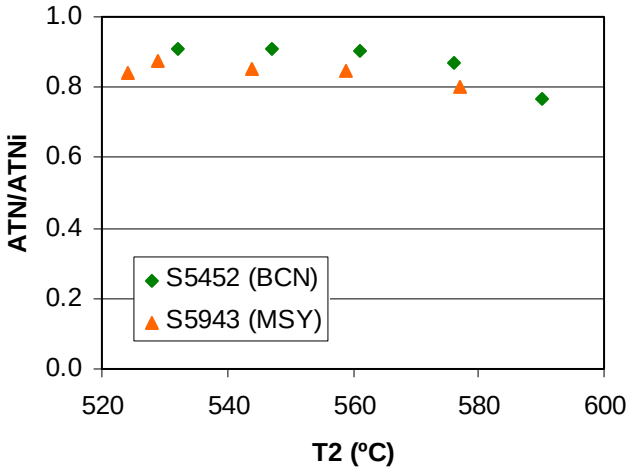

Figure S3. EC yields in the third step (ATN/ATNi), defined as the attenuation of the laser signal after stage 2 and right before stage 3 (ATN) with respect to the initial attenuation of the laser signal (ATNi) for two filters depending on the temperature during stage 2 (T2).

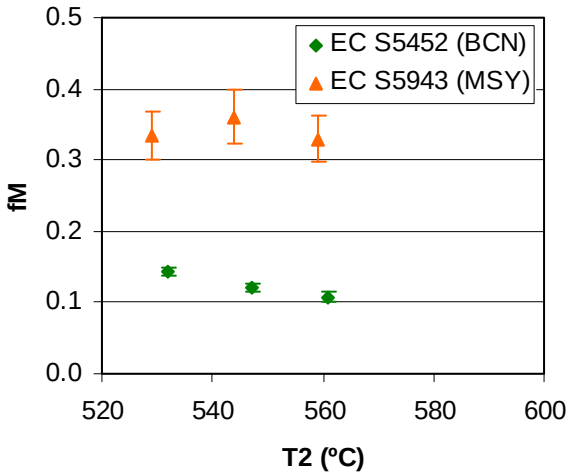

Figure S4. fM of collected CO_2 in the third step for two filters depending on the temperature during stage 2 (T2).

SECTION S5

Levoglucosan measurements

Four different data sets of levoglucosan concentrations are available, measured by four different laboratories. Levoglucosan was determined using the same filters used for ^{14}C analyses (PM_1 48h samples) by two methods: gas chromatography-mass spectrometry (Agilent 6890N gas chromatograph coupled to a mass spectrometer Agilent 5973N) by the Hungarian Academy of Sciences (lev-HAS), and by proton nuclear magnetic resonance (HNMR, Tagliavini et al., 2006) by the Institute of Atmospheric Sciences and Climate of the Italian National Research Council (lev-ISAC). A third levoglucosan dataset was analyzed by the Finish Meteorological Institute (lev-FMI) in 12h $PM_{2.5}$ samples collected on quartz fiber filters (Munktell) by high volume (30 m^3/h) samplers DIGITEL DHA-80, by high-performance anion-exchange chromatography with electrospray ionization mass spectrometry detection (Saarnio et al., 2010). Finally, a fourth levoglucosan dataset was analyzed in 12h PM_1 samples collected on quartz fiber filters (Munktell) by high volume (30 m^3/h) samplers DIGITEL DHA-80, by gas chromatography coupled to mass spectrometry (van Drooge et al., 2009; van Drooge and Ballesta, 2010) by the Institute of Environmental Assessment and Water Research (lev-IDAEA). When simultaneously available, the different levoglucosan measurements agree reasonably well (Figure S5), especially those analyzed in the same filters (lev-ISAC and lev-HAS). The highest concentrations are reported by FMI, which can be partially due to the coarser fraction ($PM_{2.5}$) analyzed. In this work, we used the average of the concentrations of lev-HAS and lev-ISAC as they are consistent with most of the measurements, and for maximum overlap with the ^{14}C dataset.

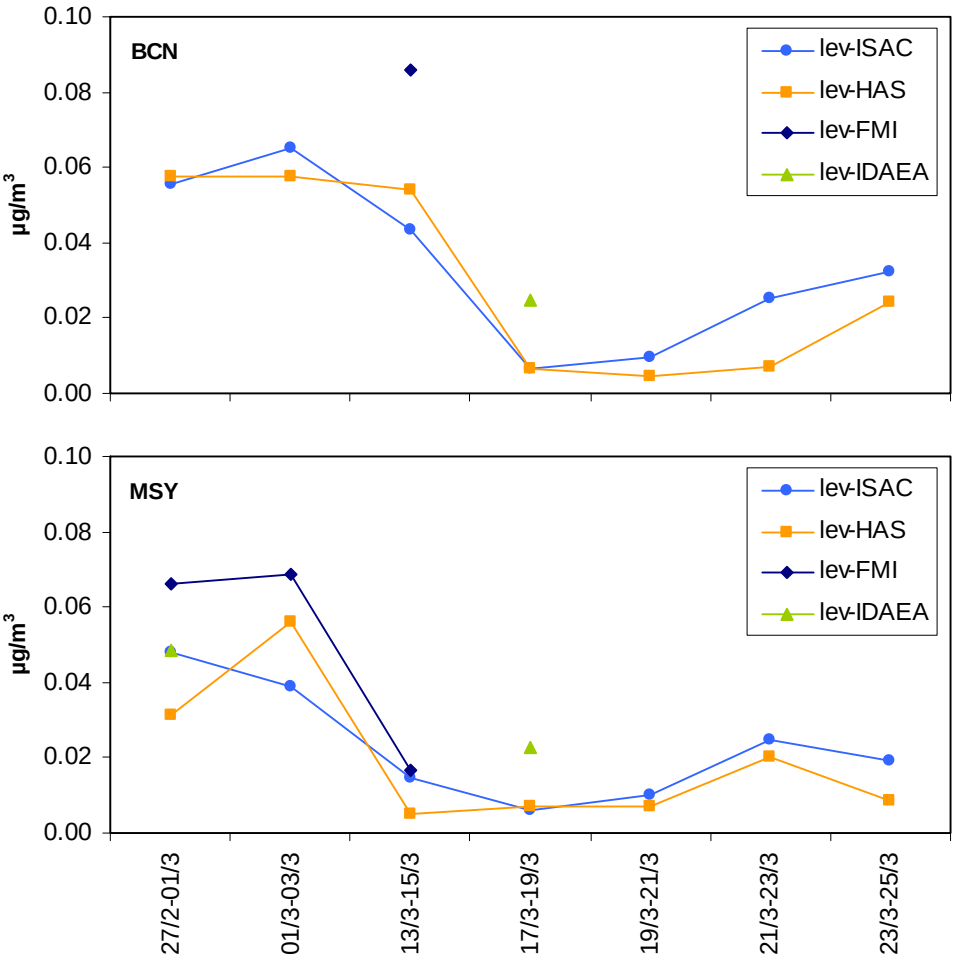

Figure S5. Levoglucosan concentrations determined by four different laboratories (see text for details). When the samples analyzed had a different time-resolution than the ^{14}C sampling periods (48h), averages of the corresponding samples are shown to match the 48h sampling periods. Only data matching the ^{14}C dataset period are shown, although more data from lev-FMI and lev-IDAEA are available.

References from Section S5:

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SECTION S6

Application of PMF to the MSY AMS data set

Positive Matrix Factorization (PMF) was performed on the organic *m/z* vs. time matrix using the PMF2.exe program and analyzed with the PMF Evaluation Tool (PET, http://tinyurl.com/PMF-guide) according to the procedures described on Ulbrich et al. (2009). 771 resolvable ions were fitted in the high-resolution analysis software, PIKA, for *m/z* 10-206, and were used in the PMF analysis. Solutions with p = 1 to 12 factors were computed. Rotational parameters (FPEAK) values between -1 and 1 were explored. An 8-factor solution (FPEAK=0) was used since this was the lowest number of factors at which the HOA and BBOA factors showed a clear separation from each other and from OOA. FPEAK=0 was used because this solution yielded the lowest Q value and the factor spectra were very similar for other FPEAK values. Six factors were recombined to make up the OOA factor since separately some of the factor spectra were not meaningful, the time series of some factor pairs had very high correlation, and other factor pairs showed signs of intermittent splitting, with one nearly disappearing when the other one appeared. No clear interpretation of the OOA subfactors was possible, as is sometimes the case in rural / remote datasets (Jimenez et al., 2009). The BBOA and HOA factors used here are those from the 8-factor solution. On average, OOA comprised 86% of the organic mass with BBOA and HOA comprising 10% and 4%, respectively. Oxygen-to-carbon (hydrogen-to-carbon) ratios for BBOA, HOA, and OOA were 0.29 (1.49), 0.09 (1.83), and 0.67 (1.37), respectively, which are consistent with other studies (e.g. Aiken et al., 2008). BBOA showed moderate to strong correlation with levoglucosan, black carbon, and total particle concentrations. HOA showed strong correlation with black carbon and total particle concentrations and moderate correlation with NO_x. The OOA factor showed correlations with black carbon and the secondary inorganic species (sulfate, nitrate, ammonium). These correlations are mostly consistent with factor identifications in other studies (Zhang et al., 2011, and references therein).

References from Section S6:

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, E. J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R.,

Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of organic aerosols in the atmosphere, Science, 326, 1525-1529, 10.1126/science.1180353, 2009.

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, Atmos. Chem. Phys., 9, 2891-2918, 2009.

Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Ulbrich, I. M., Ng, N. L., Worsnop, D. R., and Sun, Y.: Understanding Atmospheric Organic Aerosols via Factor Analysis of Aerosol Mass Spectrometry: a Review, Anal. Bioanal. Chem., DOI:10.1007/s00216-011-5355-y, 2011.

SECTION S7

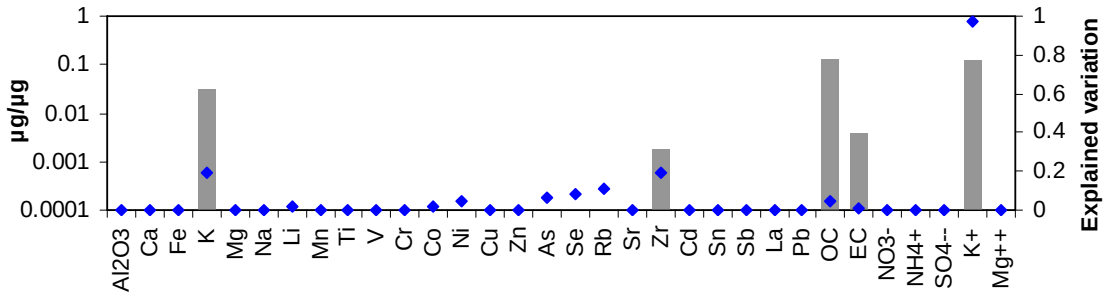
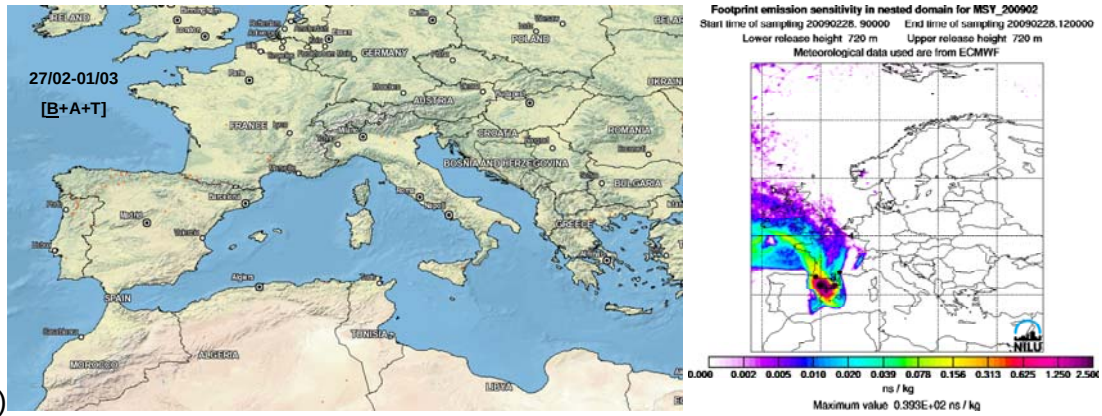


Figure S6. Source profile of the biomass burning factor and explained variation of each species by this biomass burning factor found by PMF-OF.

SECTION S8

a)



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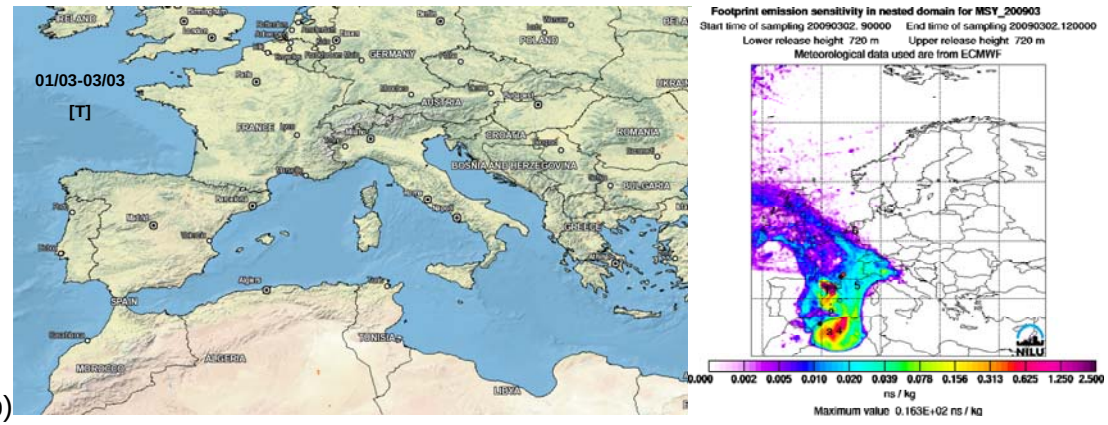

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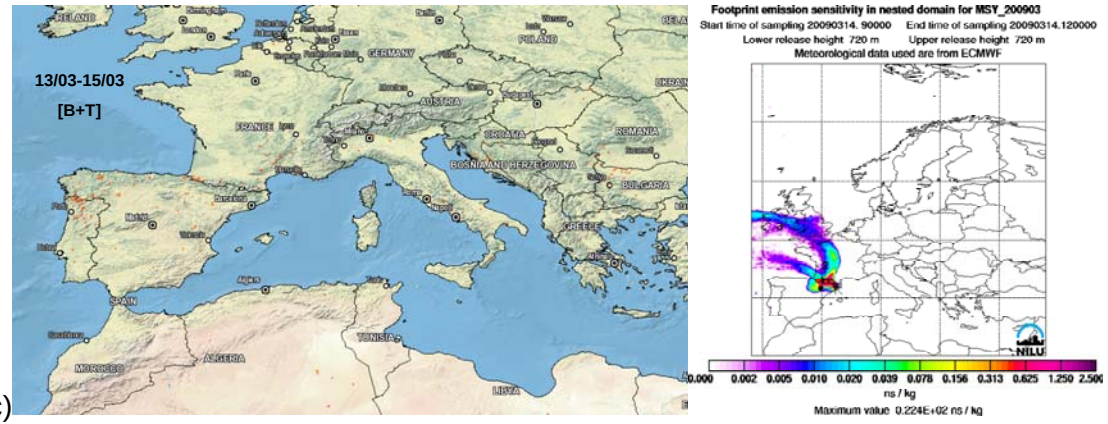
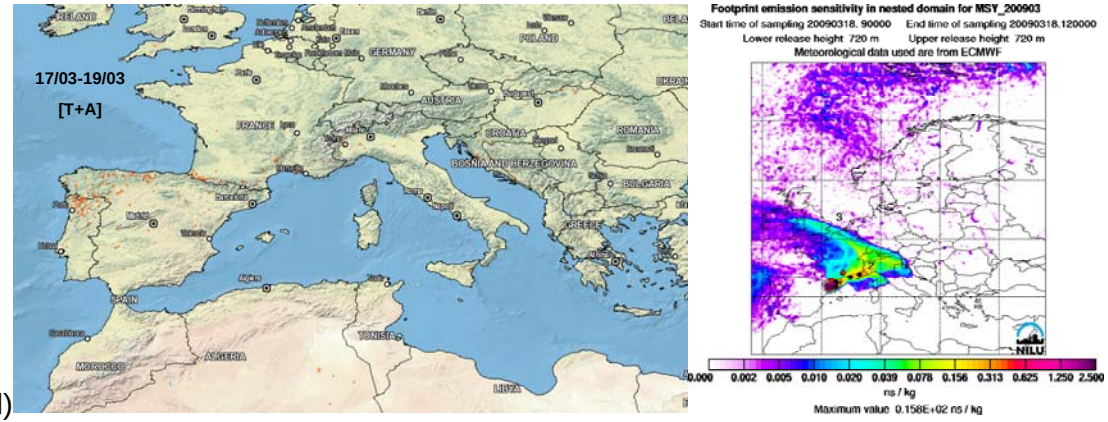
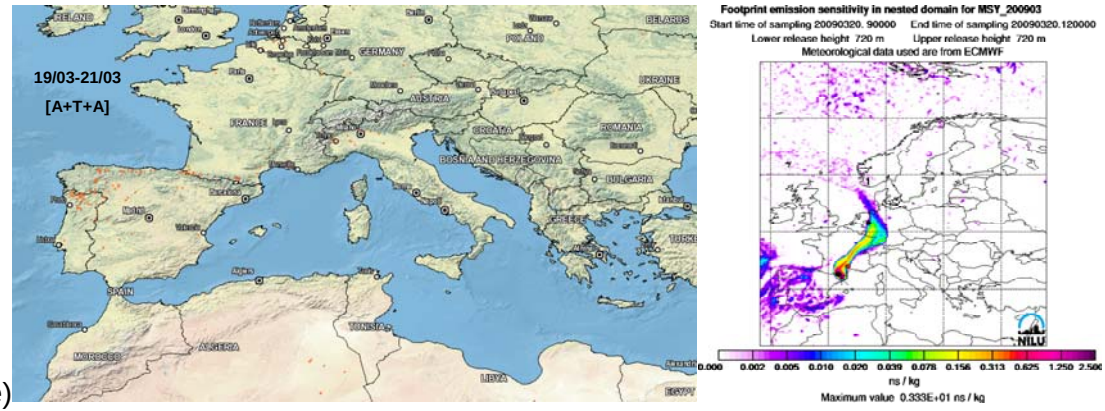


d)



e)


19/03-21/03
[A+T+A]
Footprint emission sensitivity in nested domain for MSY_200903
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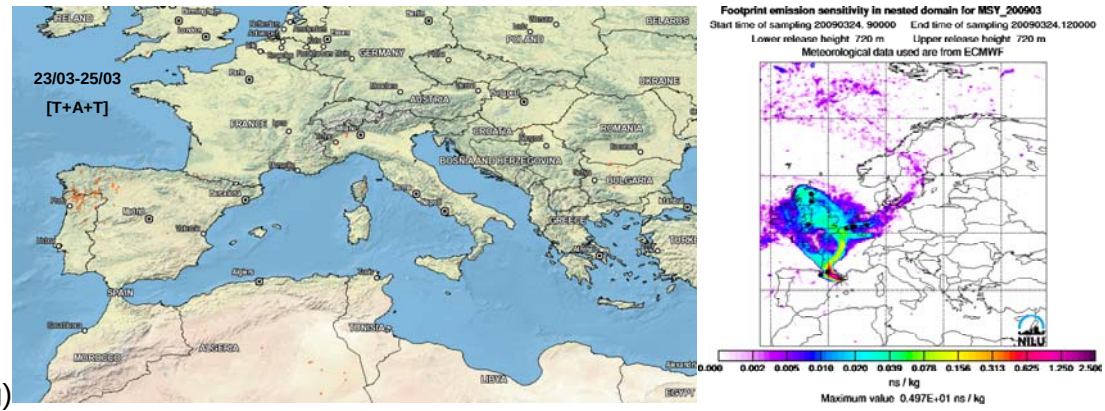
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.333E+01 ns / kg

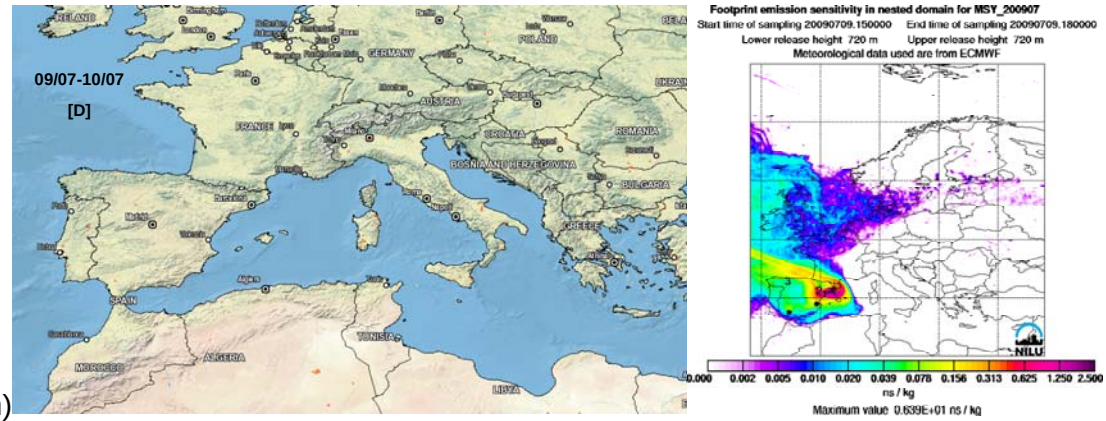
f)

21/03-23/03
[A+T]
Footprint emission sensitivity in nested domain for MSY_200903
Start time of sampling 20090322. 90000 End time of sampling 20090322.120000
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.659E+01 ns / kg

g)


23/03-25/03
[T+A+T]
Footprint emission sensitivity in nested domain for MSY_200903
Start time of sampling 20090324. 90000 End time of sampling 20090324.120000
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.497E+01 ns / kg

h)


09/07-10/07
[D]
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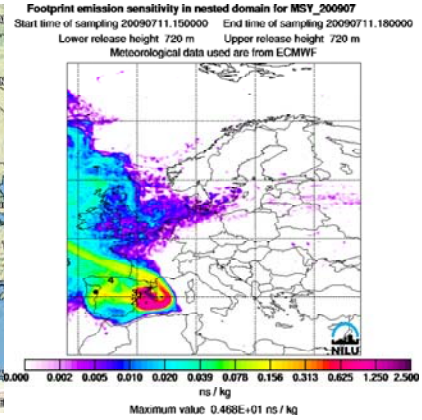
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.639E+01 ns / kg

i)

11/07-12/07
[D]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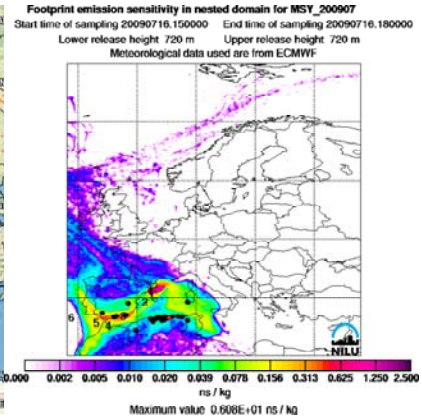
Start time of sampling 20090711.150000 End time of sampling 20090711.180000
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.468E+01 ns / kg

j)

16/07-17/07
[F]


Footprint emission sensitivity in nested domain for MSY_200907
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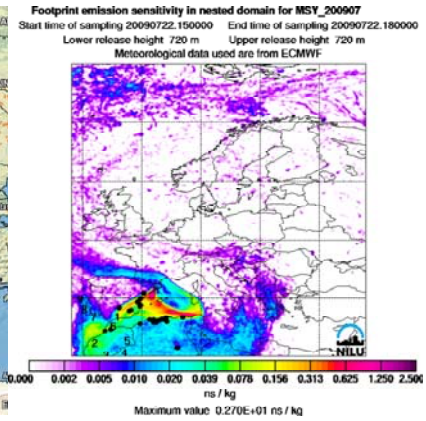
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.608E+01 ns / kg

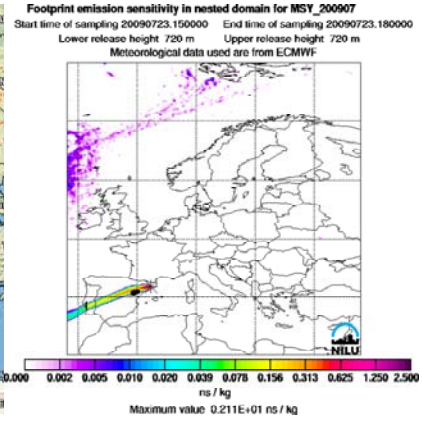
k)

22/07-23/07
[F]


Footprint emission sensitivity in nested domain for MSY_200907
Start time of sampling 20090722.150000 End time of sampling 20090722.180000
Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.270E+01 ns / kg

l)

23/07-24/07
[E+D]


Footprint emission sensitivity in nested domain for MSY_200907
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Lower release height 720 m Upper release height 720 m
Meteorological data used are from ECMWF
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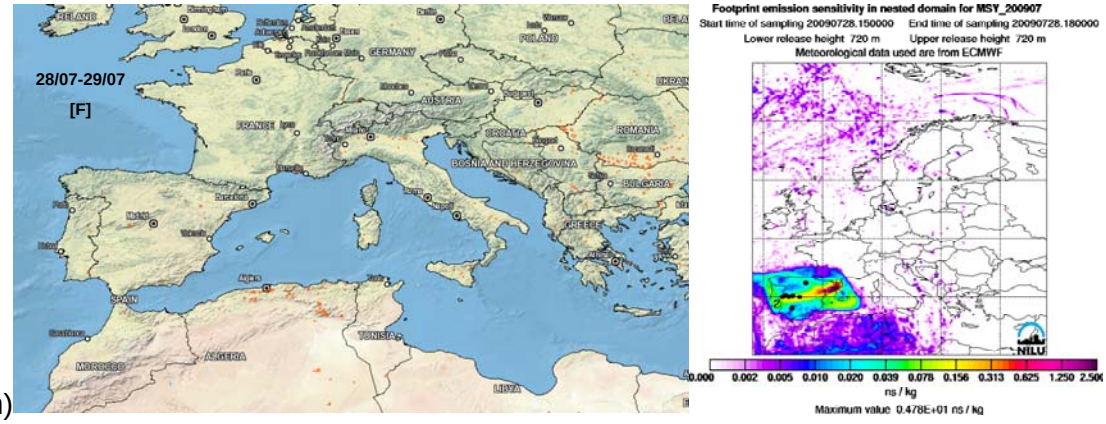
ns / kg
Maximum value 0.211E+01 ns / kg

m)


28/07-29/07
[F]
Footprint emission sensitivity in nested domain for MSY_200907
Start time of sampling 20090728.150000
End time of sampling 20090728.180000
Lower release height 720 m
Upper release height 720 m
Meteorological data used are from ECMWF
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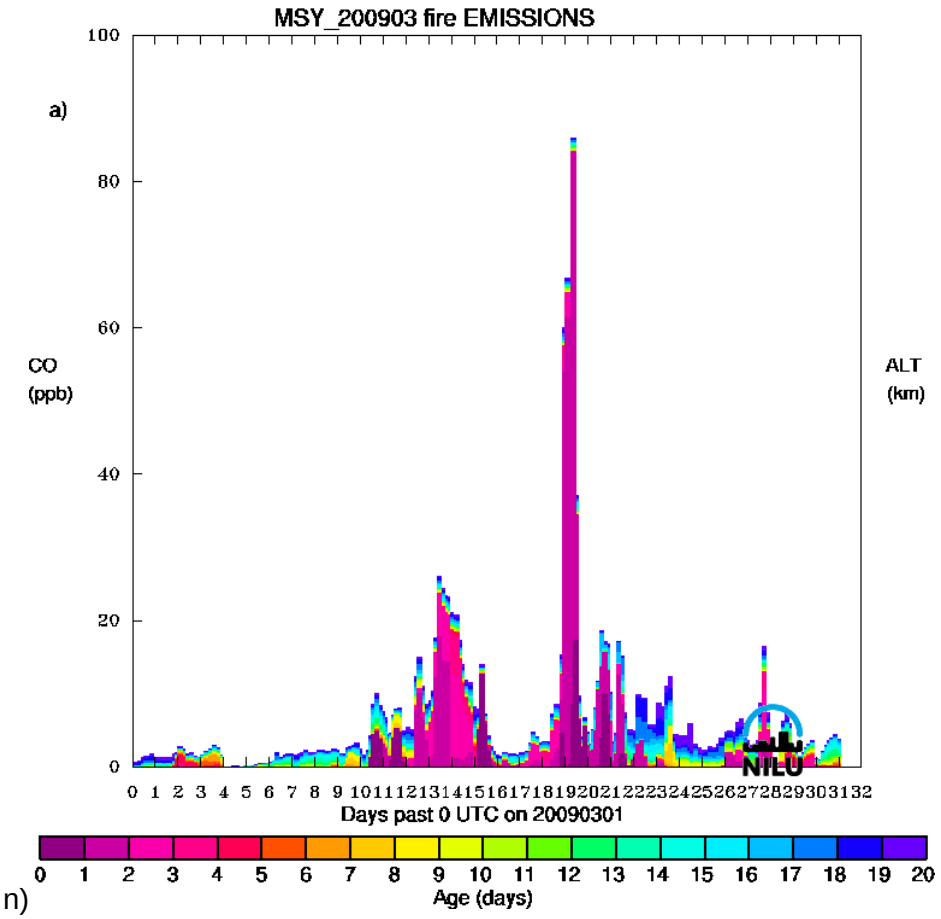
ns / kg
Maximum value 0.478E+01 ns / kg

n)


MSY_200903 fire EMISSIONS
a)
CO
(ppb)
ALT
(km)
0 20 40 60 80 100
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32
Days past 0 UTC on 20090301
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20
Age (days)

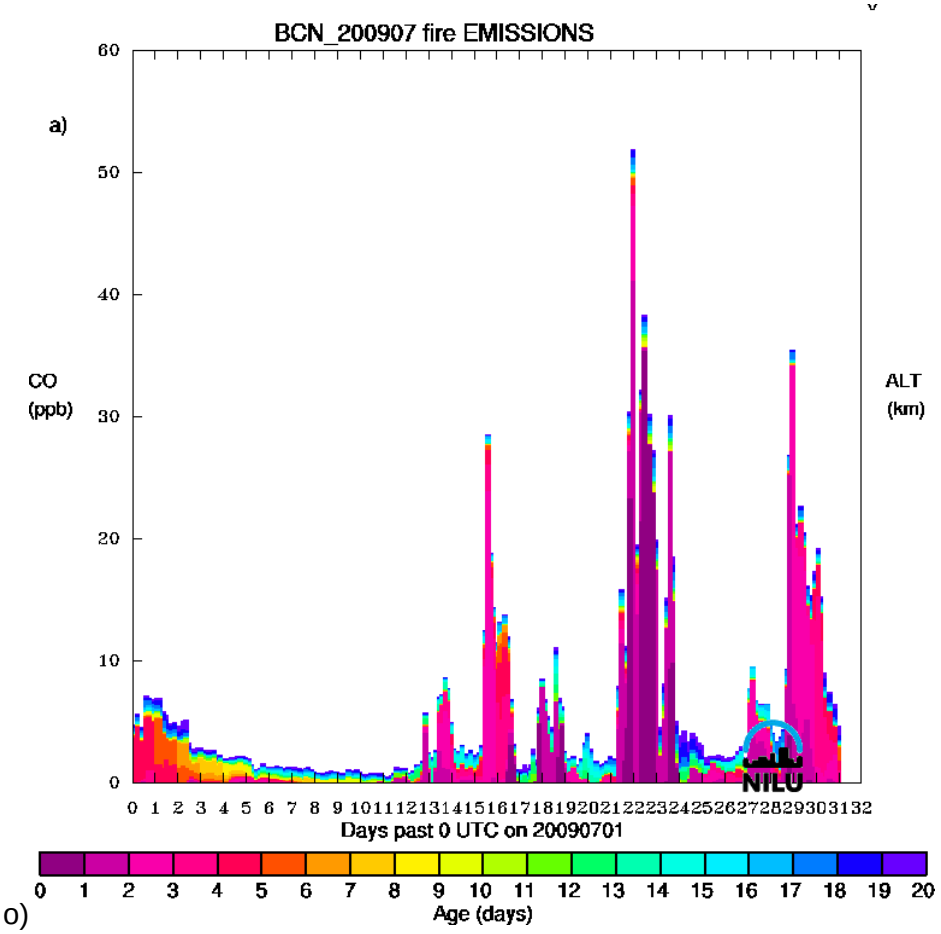


Figure S7. Hotspot/fire locations detected by the MODIS Rapid Response System provided by Web Fire Mapper, Fire Information for Resource Management System (FIRMS, Justice et al., 2002). Each map corresponds to a ^{14}C sample period (a-g: DAURE-W; h-m: DAURE-S). The atmospheric scenarios are listed below the dates; the prevailing scenario is underlined when more than one occurred during the same sampling period. FLEXPART air-mass sensitivities (Stohl et al., 2005) for MSY calculated for the middle of the sampling period. Wild fires impact estimated with FLEXPART during (n) DAURE-W and (o) DAURE-S.

References from Figure S7:

Justice, C.O., Giglio, L., Korontzi, S., Owens, J., Morisette, J.T., Roy, D., Descloitres, J., Alleaume, S., Petitcolin, F., and Kaufman, Y.: The MODIS fire products, Remote Sensing of Environment 83, 244-262, 2002.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461-2474, 2005

SECTION S9

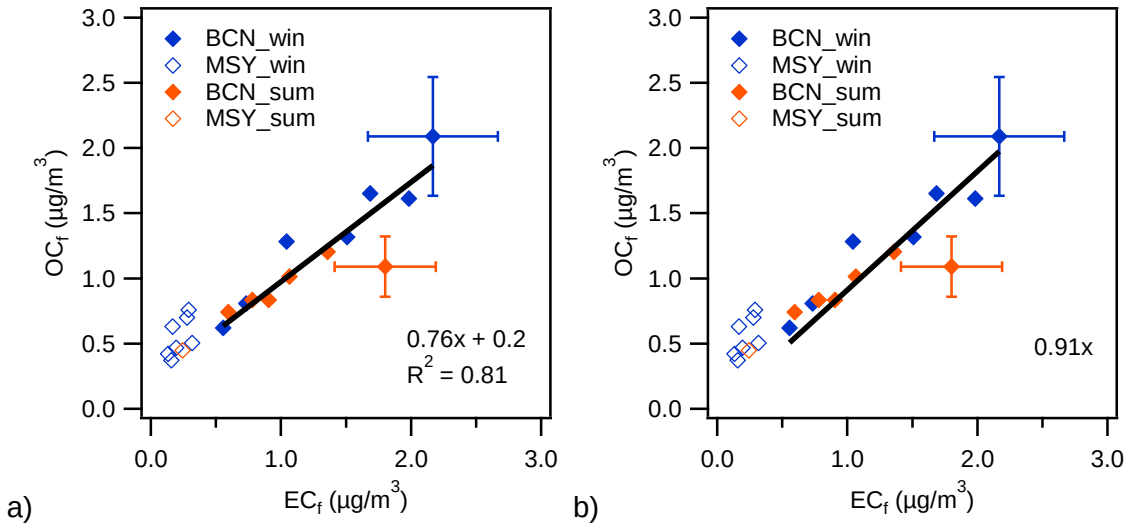


Figure S8. OC_f vs EC_f concentrations at BCN and MSY during DAURE-W and DAURE-S. Error bars indicate measurement uncertainty, only shown for two data points for clarity. Line and equation correspond to orthogonal distance regression for BCN. a) Regression line allowing intercept. b) Regression line with intercept forced to zero.

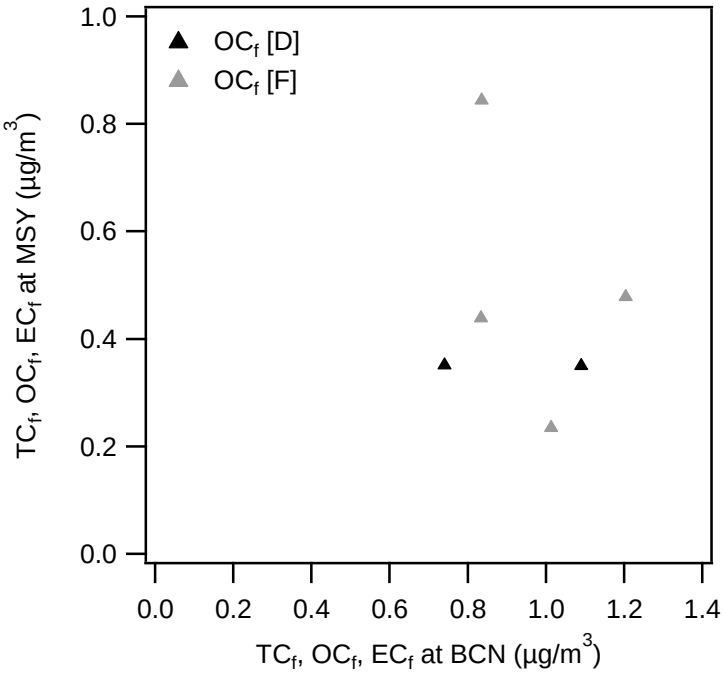


Figure S9. OC_f contributions at MSY vs OC_f contributions at BCN during scenario [D] and during scenario [F].